

GUIDANCE FOR DECONTAMINATION OF WATER SYSTEM INFRASTRUCTURE

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INTRODUCTION

The objective of this project was to develop and test protocols for decontamination of pipe surfaces. The intent was to develop an approach and methodology that other laboratories could use to generate comparable data. Because of the vast number of potential contaminants, the numerous water quality conditions, and variety of pipe materials that could exist within distribution systems, it was not possible to anticipate all situations where decontamination of water utility infrastructure would be necessary. Rather, the experimental approach outlined a framework for evaluating the impact of various parameters to estimate their importance on the selection of decontamination procedures. This framework should nevertheless be useful to all water utilities and decision makers in selecting appropriate actions during emergencies. Moreover, the work will serve as a basis for additional investigation for the development of detailed SOPs and specifically tailored cleaner formulations.

The experimental approach was divided into three phases: 1) an initial evaluation to examine the impact of water matrix parameters (temperature, pH, alkalinity, total organic carbon [TOC]) testing, 2) the impact of pipe material on the adherence of contaminants to pipe surfaces, and 3) examination of various agents to enhance the removal of contaminants from the pipe surfaces. Because of the numerous combinations of water quality conditions, pipe materials, and decontamination agents, it was necessary to conduct the experiments in phases to reduce the number of variables. This approach undoubtedly left some important informational gaps that will require additional research. This framework approach, however, was necessary to identify the important variables to help focus future research and to aid decision makers in the absence of more definitive information.

Contaminant Application Selection

The initial decisions focused on the selection of contaminant agents. Bacterial spores and bacteriophages were selected as surrogates for microbial contaminants. *Bacillus thuringiensis* subsp. *israelensis* spores were selected as a surrogate for *Bacillus anthracis* spores, the causative

agent of anthrax. *Bacillus thuringiensis* is genetically very similar to *B. anthracis* and is generally considered a good surrogate for treatment applications (Rice et al. 2005). MS-2 is commonly used as a surrogate for Poliovirus and other human enteric viruses, and was selected because its high affinity for surfaces (LeChevallier and Au, 2004). Vegetative bacterial cells were not examined because these organisms were considered to be easily disinfected and conditions adequate to control bacterial spores would be sufficient for inactivation of vegetative bacteria. Protozoan cysts such as *Cryptosporidium parvum* oocysts were not examined because of the difficulty of the analytical methods, although *Bacillus* spores have been suggested as a surrogate for *Cryptosporidium* treatment (Lytle et al. 1999). Several inorganic compounds were selected because they are toxic substances (mercuric, arsenate, and cyanide,) and are obvious threats to water. Other inorganic compounds were selected as non-radioactive surrogates for their radioactive isotopes (salts of cobalt, strontium, cesium, and thallium) although some of these non-radioactive compounds may also have public health consequences. Organic contaminants can be characterized based on their octanol/water partition coefficient (indicated as $\log_{10} P_{ow}$ or simply $\log P$ or $\log K_{ow}$.) which quantifies the hydrophobic characteristics of the compound. Chemicals with high $\log P$ values are hydrophobic and tend to bind readily to surfaces, while those with low $\log P$ values are hydrophilic and are more soluble in water. Chlordane was selected as a high $\log P$ chemical surrogate with a $\log P$ value of 6.2, while *p*-dichlorobenzene (*p*-DCB) was selected as a moderate $\log P$ surrogate with a $\log P$ value of 3.4. *p*-Dichlorobenzene is the ingredient used in some moth balls; while chlordane was used as a foundation treatment for termites (it is no longer available in the US). For all these contaminants decisions had to be made regarding the concentration to be used. The concentration had to be high enough to impart a substantial residual on the pipe surface (for subsequent decontamination), yet not so concentrated as to precipitate in stock solutions or to interact with other contaminants so that quantitative recovery and calculation of mass balances could not be performed.

In devising the experimental procedures, other decisions had to be made regarding the water quality matrix. It was considered likely that parameters such as temperature, pH, alkalinity, and TOC would likely affect the interaction of the contaminants with the pipe surfaces. Some effects, however, could be counterintuitive. For example, low temperatures could make some compounds more viscous and potentially adhere more readily to surfaces. Additionally, lower temperatures would reduce chemical reaction rates with pipe surfaces and potentially decrease attachment. The pH of the water samples could have various effects on the solubility or reactivity of a compound, depending on its carbonate chemistry (carbonate frequently controls the solubility of many inorganic compounds in water) and interactions with organic ligands. The type of natural organic matter in the water can have profound impacts on the interaction of organic, inorganic, and microbial contaminants that can either increase or decrease the attachment with pipe surfaces. Because it was not practical to examine all types of organic matter that may be present in water, it was decided to use a standard humic acid preparation. Future research may expand on the protocol by examining other types of organic carbon to better understand the impact of this variable on decontamination procedures.

Pipe Selection

The types of pipes examined were selected to represent a range of commonly used materials in drinking water and domestic plumbing:

- | | |
|---|---|
| 1. cPVC (control) | 7. Copper |
| 2. cPVC with biofilm | 8. Cement lined ductile iron (without seal coat) |
| 3. Iron (control) | 9. Cement lined ductile iron (with asphaltic seal coat) |
| 4. Iron with biofilm | 10. Polyethylene (PE) |
| 5. Galvanized (new) | 11. Epoxy coated steel |
| 6. Used galvanized (with biofilm and heavy tuberculation) | |

The objective of the research was to determine if unique interactions occurred between various contaminants and certain materials – not to inventory a comprehensive list of distribution system materials. Some materials were incubated for several weeks to grow bacterial biofilms on the pipe surface. Comparison of the pipes with biofilms to the control un-inoculated pipe surfaces would allow an evaluation of the role of biofilms on contaminant adsorption. Research has shown that *Cryptosporidium* oocysts and some viruses can be accumulated in biofilm samples (Quignon, et al. 1997; Piriou et al., 2000) and the presence of biofilms could either increase or decrease the interaction of various contaminants (especially organic and inorganic compounds). Some heavily tuberculated galvanized iron service line pipes were obtained from a distribution system to examine the effect of this corroded surface on contaminant adsorption.

It was also decided to focus on pipeline materials since these, of all water utility infrastructure, are the most difficult to access, clean and inspect. It is recognized that other structures could become contaminated (e.g., filter material, pumps, tanks, etc.), but options exist to either replace the material (e.g., filter media) or clean the surfaces. In these cases, the cleaning procedures developed for pipe surfaces should be applicable to other types of water utility infrastructure (e.g. tanks or pumps).

Test Protocol

The exposure protocol used in this study involved filling pipe sections and incubating the pipes with the contaminants for periods up to seven days. Although the pipes were rotated on a daily basis, this static exposure simulated the contact that would occur once a system was shut down following a contamination event. An alternative exposure protocol could have used a flowing system, but this approach would have been more complicated and would have limited the number of experimental trials. Still another mechanism considered included mechanical shaking to simulate the flushing movement and pulsing movements in a distribution system. Hydraulic shear forces and currents are known to be important transport mechanisms influencing the attachment, and desorption, of particles with surfaces (Characklis and Marshall 1990, Ollos et al. 2003), but given the range of flow velocities that may occur in distribution systems, it was deemed too complicated for this project to try to generalize how these factors may influence contaminant attachment. Additional research will be necessary to evaluate these interactions.

Pipe sections (one foot) were washed by decanting the contaminant mixture and sequentially refilling the pipes with fresh wash water. An alternative approach could have used a flowing

system that flushed the pipes with various volumes and velocities of water. However, these complicated flushing studies would tell us more about principles known to be important for flushing (Friedman et al. 1998; Kirmeyer et al. 2000), rather than examining the interaction of the contaminants with the pipe surfaces.

Finally, it was necessary to make decisions on the selection of potential decontamination agents to limit the compounds to a manageable number. This study initially screened 11 surfactants and then tested 8 compounds in addition to the three standard procedures used to desorb the contaminants from the pipe surfaces (termed a “getter wash”). Clearly, there is a “universe” of other decontamination agents that could have been evaluated. The selection of decontamination agents used in this study was a feasibility analysis to see if improved detachment was possible, and to provide guidance on the types of approaches that might be useful in future studies rather than a comprehensive evaluation of all possible cleaning alternatives. More importantly, the approach used in this study provides a framework for vendors to evaluate their products and make comparisons.

Experimental Objectives

The objective for the experimental phase of the projects was to develop and evaluate a protocol for the decontamination of distribution system pipelines following an intentional contamination event. The experimental phase objectives included:

- ☐ Develop a protocol to evaluate the accumulation of contaminants on pipe surfaces.
- ☐ Evaluate the protocol using organic, inorganic and biological contaminants to determine the degree of attachment to various pipe materials.
- ☐ Evaluate conventional and novel technologies for the decontamination of pipelines from contaminants that may be accumulated on pipe surfaces.
- ☐ Develop recommendations based on the experimental evidence to guide decision makers in the event of an intentional (or unintentional) contamination of water utility infrastructure.

By necessity, this was a screening level study that examined a variety of pipes, water chemistry variables, and decontamination approaches under laboratory (static) conditions. Much more specific research will be necessary to elaborate on these initial findings and refine the suggested SOPs under a range of real world conditions.

MATERIALS AND METHODS

Source of Materials

The *Bacillus thuringiensis* subsp. *israelensis* (ATCC 35646) stock was provided by Dr. Eugene Rice of the USEPA in Cincinnati, OH. The MS2 bacteriophage was obtained from the American Type Culture Collection (ATCC 15597-B1). The inorganic chemical contaminants including: sodium cyanide (Lot #F19M04), sodium hydrogen arsenate heptahydrate (Lot#D04N03), strontium chloride hexahydrate (Lot#F20N18), cobalt chloride hexahydrate (Lot#E19N06), and thallium chloride (Lot#J10N29) were obtained from Alfa Aesar (Ward Hill, MA). Mercuric acetate (Lot#Y51608) was purchased from J.T. Baker (Phillipsburg, NJ) and cesium chloride (Lot#B48328) was purchased from Calbiochem (San Diego, CA).

Chlordane is no longer manufactured in the United States and a pint of commercial blend (Chevron Ortho-Klor 74, chlordane spray, active ingredients: 74% technical chlordane [Octachloro-4,7-methanotetrahydroindane and 29.6% related compounds] and 21% petroleum distillate; inert ingredients 5%) was obtained from a toxic waste facility in Fairfax County, VA). Paradichlorobenzene (p-DCB, Lot#44114423) was obtained from EM Science (Gibbstown, NJ).

Contaminant Analysis

Appendix 1 contains the detailed protocol developed during the water matrix testing (Phase 1a). The baseline formulation of the artificial water matrix for all experiments was pH 7.2, alkalinity of 100 mg/L NaHCO₃ and 20 mg/L CaCO₃, and 2 mg/L TOC (from humic acid). The biological contaminant mixture consisted of *Bacillus thuringiensis* (a spore-forming organism related to the anthrax organism *B. anthracis*) and the bacteriophage MS2, both at approximately 10⁴-10⁵ viable units per mL. The two microbes were mixed together and selectively plated using procedures outlined in Appendix 1. In the decontamination method (Phase 2), the biological contaminant mixture consisted only of *Bacillus thuringiensis* at approximately 10⁴-10⁵ viable units per mL.

Initially, the inorganic contaminant stock solutions were prepared at 1 g/L (as product). However, this high concentration resulted in precipitation problems upon storage so the concentrations were reduced to 0.1 g/L with minimal problems. To avoid chemical interactions, and to minimize interferences in analysis, the inorganic contaminants were prepared in four mixtures:

1. Sodium cyanide was prepared separately.
2. Sodium hydrogen arsenate heptahydrate, strontium chloride hexahydrate, and cesium chloride were prepared as a mixture.
3. Mercuric acetate was initially included in the arsenic-strontium-cesium mixture, but was eliminated during the water matrix testing to avoid precipitation. Mercuric acetate was prepared separately in the adherence to pipe substrate assays (Phase 1b).
4. Cobalt chloride hexahydrate and thallium chloride were prepared as a mixture.

All inorganic analyses were performed at the National Environmental Laboratory Accreditation Conference (NELAC) certified, American Water Quality Control Laboratory in Belleville, Illinois. Cyanide was analyzed using EPA Method 335.4 Rev 1.0 and mercury by EPA Method 245.2 (Cold Vapor Atomic Absorption). Arsenic, Strontium, Cesium, Cobalt and Thallium were determined using EPA Method 200.8 Rev 5.4 (ICPMS).

The two organic compounds, chlordane and *p*-dichlorobenzene (p-DCB), were prepared as a mixture and were analyzed by EPA Method 525.2 Rev 2.0 (Semi-volatiles). The concentration of chlordane was calculated as the average of chlordane (gamma chlordane, alpha chlordane and trans nonachlor). The reporting limits for the contaminants were as follows:

Analyte	Reporting Limit
<i>Bacillus</i>	1 colony forming unit
MS2	1 plaque forming unit
Cyanide	0.005 ppm
As	0.001 ppm
Co	0.010 ppm
Cs	0.001 ppm
Hg	0.0002 ppm
Sr	0.050 ppm
Tl	0.0003 ppm
p-DCB	0.5 ppb
Chlordane	0.1 ppb

Appendix x provides quality control results for the analytical methods.

Pipe Materials

The adsorption of microbial, inorganic, and organic contaminants was tested using the following pipe materials:

1. cPVC (control)
2. cPVC with biofilm
3. Iron (control)
4. Iron with biofilm
5. Galvanized (new)
6. Used galvanized (with biofilm and heavy tuberculation)
7. Copper
8. Cement lined ductile iron (without seal coat)
9. Cement lined ductile iron (with asphaltic)



Figure 1. Pipe materials examined (from left to right): cPVC, cPVC with biofilm, black iron, black iron with biofilm, used galvanized, new galvanized, copper, polyethylene, epoxy coated steel, cement lined ductile iron pipe without seal coat, cement lined ductile iron pipe with seal coat, cement lined ductile iron pipe with asphaltic

seal coat)

11. Epoxy coated steel

10. Polyethylene (PE)

The polyvinylchloride (PVC, grade “c” ASTM D-2846), black iron, and (new) galvanized pipes were purchased from a local hardware store as nominal 0.75, 0.5, and 1.0 inch diameter stock, respectively. Copper pipe (Type 1" L, NSF-61, 04831; nominal 1.0 inch diameter) was obtained from Howell Metal Company, (New Market, VA). The cement-lined ductile iron pipe was provided both with and without an asphaltic bituminous seal coat (International B-523, International Oil) from the manufacturer (McWane Cast Iron Pipe, Birmingham, AL; nominal 3.0 inch diameter). The polyethylene (stock #3408, Orangeburg Poly-Xtra, conforming to ASTM D2239) and epoxy-coated steel pipes (APL-2000, American Pipe Lining, San Diego, CA), both nominal 1.0 inch diameter, were provided by the Civil Engineering Research Laboratory (CERL) of the Army Corps of Engineers in Champaign, Illinois. Heavily corroded, used galvanized service line pipe (nominal 1.0 inch diameter) was obtained from a participating water utility. Biofilms were grown on cPVC, iron, and the tuberculated used galvanized pipes by incubating the pipes with the synthetic water (containing 2 mg/L TOC) for 2 weeks in the lab.

Experimental Protocol

During the water matrix testing and adherence to pipe substrates, the general procedure involved filling one-foot pipe sections with the contaminant mixtures and allowing the contaminants to interact with the pipe surface for 7 days. The pipe sections were sealed using various cap materials (rubber stoppers or parafilm) and rotated (top to bottom) on a daily basis to ensure even contact with the pipe surface. After the incubation period, the pipes are drained, the caps replaced, and the pipes were refilled with the wash water (no contaminants) and the pipe surfaces were rinsed a total of three times (two rinses and a “getter wash”). The final rinse was performed using a more aggressive procedure (termed a “getter wash”) to try to remove any remaining contaminant that might still be adsorbed to the pipe surface. The contaminant concentration in the stock solution, the water after the 7 day incubation, and in each of the three washes were analyzed and the data expressed as both the concentration of contaminant in solution (in units per liter) or in total mass within the pipe vessel. The data were analyzed to determine the total amount of contaminant adsorbed to the pipe surface and the amounts released in the subsequent washes.

Different configurations of the “getter wash” were utilized depending on the type of contaminant analyzed. The final “getter” wash for the microbial contaminants was one pipe volume of phosphate buffered saline and the pipe surface was scrubbed using a test tube brush. The getter wash for the metal contaminants was 0.1 M ammonium chloride, and final decontamination experiments exposed the 0.1M ammonium chloride to the pipe for 24 hours. Initially, the getter wash for both organic contaminants was a 50% methanol solution, but subsequent experiments showed that solubility was improved using a 100% methanol solution. The higher methanol concentration had a slight negative impact on the efficacy of the semi-volatile analysis (Method 200.8 Rev 5.4), reducing the recovery by about 10% (see Appendix x), but the change in efficacy was accounted for by analyzing controls. For testing the water matrix and pipe material effects, the surfaces were simply rinsed with 100% methanol and decanted. For the decontamination studies, the 100% methanol solution was held for a one hour exposure on organic contaminated pipes.

Decontamination Procedure

During the examination of decontamination procedures, the experimental protocol was slightly modified so that the decontamination agent was used in place of the first wash. The decontamination agent was applied to the contaminated test pipes for 24 hours (Figure 2). The pipe surfaces were then rinsed with one wash water and a final “getter” wash. For the chlordane and p-DCB assay with the used galvanized pipe, the pipe was rinsed three times (before the final getter wash) after the decontamination step to avoid the carry-over of the surfactants into the organic assay. For all assays, the effectiveness of the decontamination procedures was calculated by determining the amount of contaminant in the final getter wash compared to the final getter wash from non-decontaminated pipes. If the decontamination agent was effective in removing the contaminant, the contaminant in the final wash would be much lower than the control where water washes alone were used.

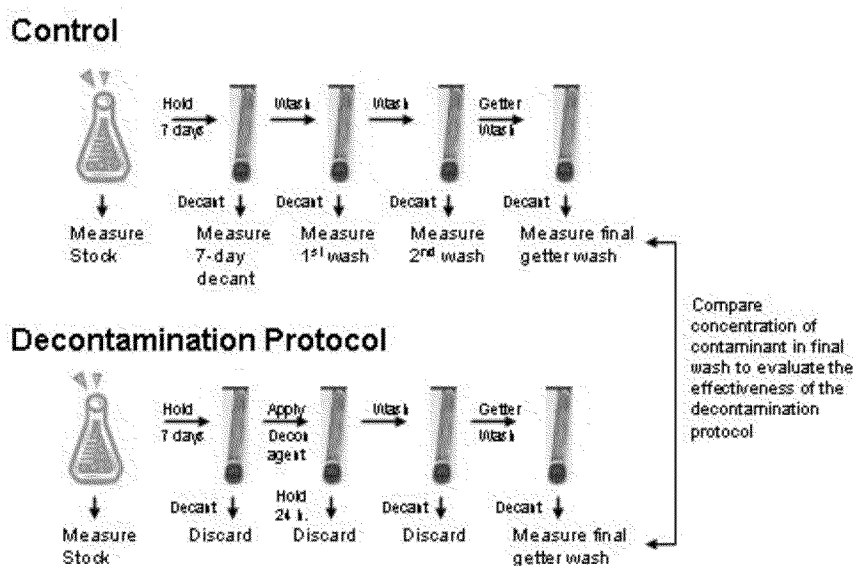


Figure 2. Schematic of the decontamination protocol.

To evaluate the effectiveness of the decontamination procedure, the concentration of remaining contaminant in final “getter” wash was compared to a control (without the decontamination treatment).

Eight different chemicals were tested as potential decontamination agents (Table 1). Chlorine was chosen as a biocide for microbial contaminants, and EDTA and sodium citrate were selected as chelating agents for inorganic compounds. Simple Green is a commercially available cleaner that has both detergents and chelating agents. Three surfactants were selected after initial screening for treatment of organic compounds. RE-Ox is a proprietary commercial pipe cleaner that was tested for scale removal in the heavily tuberculated galvanized iron pipe.

Table 1. Selected decontaminant agents.

Decontamination Agent	Vendor	Lot #	Vendor Location
Ultra Clorox Simple Green	The Clorox Company Sunshine Makers, Inc.	E643372 19 MD2 VER: 13005A04 PW5119077	Oakland, CA Huntington Harbour, CA
Na EDTA	EMD Chemicals, Inc.	45146525	Gibbstown, NJ
Na Citrate	EMD Chemicals, Inc.	GC4D111K5D	Gibbstown, NJ
Surfonic N-60	Huntsman Petrochemical Corporation	4Q-7	Houston, TX
Surfonic TDA-6	Huntsman Petrochemical Corporation	T9464a14a	Houston, TX
Empicol LZV	Huntsman Petrochemical Corporation	140419	Houston, TX
RE-Ox	RE-Ox LLC		Las Vegas, NV

Initially a larger suite of surfactants (Table 2) were screened for their ability to dissolve chlordane. Briefly, one drop of chlordane and one drop of surfactant was added to 15 mL of synthetic water and shaken for 1 minute. The tubes were left to settle for a brief interval, observed, and then reshaken for an additional 1 minute. A control of surfactant alone in 15 mL of synthetic water was used for comparison. The tubes were scored for physical changes in color, clarity, foam intensity, and transformation (dissolved status). The experiment was repeated using a final concentration of 2.5% surfactant and 1.5% chlordane. Based on the ability to alter the physical appearance of the chlordane, Surfonic N-60, Surfonic TDA-6, and Empicol LZV were selected for testing in the pipe decontamination protocol.

Table 2. Surfactants initially screened for treatment of organic contaminants.

Surfactant	Vendor
Surfonic N-60 (nonionic surfactant), nonylphenol ethoxylate	Huntsman Petrochemical Corporation, Houston, TX
Surfonic TDA-6 (agricultural emulsifier), ethoxylated tridecyl alcohol	Huntsman Petrochemical Corporation, Houston, TX
Surfonic PE 1198 (anionic surfactant)	Huntsman Petrochemical Corporation, Houston, TX
POG 600, polyethylene glycol	Huntsman Petrochemical Corporation, Houston, TX
Surfonic POA-17R2, polyol	Huntsman Petrochemical Corporation, Houston, TX
Surfonic L24-5, C12-C16 alcohol +5 mole ethoxylate	Huntsman Petrochemical Corporation, Houston, TX
Surfonic N40, nonylphenol ethoxylate	Huntsman Petrochemical Corporation, Houston, TX
Chemco Super D, citrus terpene	Chemco Products Inc., Howell, MI
Crystal Simple Green Degreaser	Sunshine Makers, Inc., Huntington Harbour, CA
Empicol LXV100/E (solid detergent), C10-C16 sulfate sodium salt	Huntsman Petrochemical Corporation, Houston, TX
Empicol LZV (solid detergent), C12-C18 sulfate sodium salt	Huntsman Petrochemical Corporation, Houston, TX

Method Performance - Mass Balance Experiments

To determine the effectiveness of the analytical methods and experiment procedures, total mass balances were calculated to account for all the contaminants added to the pipe sections. The total

mass balance was determined by comparing the sum of the contaminant mass from the pipe sections (decanted concentrations after the 7 day incubation, each of the washes, and the final getter wash) to the mass of the contaminant initially added to the pipe section. The percent recovery was calculated as the total mass of the contaminant recovered from the pipe divided by the mass added to the pipe, multiplied by 100. Because these calculations were the result of multiple analyses, it was considered acceptable to have variations of $\pm 20\%$.

The recovery efficiency of contaminants varied depending on the pipe material. For example, mercury was recovered at 117% from cement-lined ductile iron pipes with a seal-coat, but only 47% from cement-lined ductile iron pipe without a seal-coating (Table 3). Some of the reduced efficiency was assumed to be related to chemical changes upon storage (Table 4) and attempts to control the pH changes were not successful even with preconditioning of the pipe materials (for 30 days) or use of a stronger buffer (Table 5). Some agents, like MS2 did not survive at high pH levels, whereas other agents, like cyanide, were assumed to be reactive with some pipe materials. Most of the contaminants had adequate mass balance recoveries (75 to 125%) from non-reactive materials like cPVC pipe. The major exception was p-DCB which had an analytical recovery efficiency of approximately 50%, presumably due to interferences from methanol in solution. The initial recovery efficiency of *Bacillus* spores averaged 107% (n=8) and 80% (n=6) for cPVC and iron pipe, respectively, but this efficiency unexpectedly dropped in subsequent experiments, possibly due to pH or toxic effects from some pipe materials (e.g., copper or zinc), but was unexplained in un-reactive materials such as PVC or polyethylene.

Table 3. Percent recovery of contaminants from various pipe materials.

Contaminant	cPVC	cPVC- biofilm	Fe	Fe- biofilm	New Galvanized	Used Galvanized*	Cu	Cement*	Cement w/seal*	PE	Epoxy*
Cyanide	80.9	13.9	27.0	20.0	47.4	0.5	0.1	88.7	54.3	100.3	56.6
Arsenic	117.4	108.7	31.5	8.3	42.9	11.5	118.3	26.0	105.9	110.4	118.4
Cesium	117.5	109.9	82.5	106.5	87.7	120.5	138.2	42.5	120.5	128.9	137.6
Strontium	120.2	108.9	86.7	40.5	81.4	35.3	118.9	60.0	117.0	110.1	116.5
Mercury	120.1	90.6	27.6	87.1	11.2	65.5	19.4	46.7	103.1	79.9	38.4
Cobalt	99.6	113.5	16.1	36.1	25.2	60.4	114.1	80.9	8.2	95.4	81.6
Thallium	101.8	112.5	80.1	83.0	53.0	9.9	121.1	5.8	2.0	107.1	103.4
Chlordane	74.9	59.4	49.0	75.5	47.2	67.9	62.3	43.5	50.2	37.3	43.6
p-DCB	13.2	14.8	31.2	47.8	50.7	48.9	20.1	40.5	25.5	2.8	24.4
Bacillus ^	27.0	16.0	4.0	19.0	3.0	2.0	1.0	2.0	6.0	2.0	114.0
MS2 ^	57.0	31.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	56.0	85.0

^ Measured using the stock value

*Used galvanized (with biofilm and heavy tuberculation), cement lined ductile iron (w/o seal coat), cement lined ductile iron (with seal coat), and epoxy coated steel pipe.

Some of the variability in mass balance recovery efficiencies were due to analytical methods, manipulations of the pipe samples, and specific interactions with pipe materials.

Table 4. Effect of pipe material on the pH of the test water*

Substrate	Day 0 pH	Day 7 pH
cPVC	7.5	7.2
Iron	7.5	9.7
New galvanized	7.5	9.3
Used galvanized	7.5	7.4
Copper	7.5	7.3
Cement lined ductile iron (w/o seal coat)	7.5	11.4
Cement lined ductile iron (with seal coat)	7.5	10.9
Polyethylene	7.5	7.4
Epoxy coated steel	7.8	7.7

* Synthetic water of 100/20 mg/L of bicarbonate and carbonate and all experiments used the same dihydrogen and monohydrogen phosphate concentration (see Appendix 1).

A 28 day pre-exposure to tap water (from the New Jersey American Water Delaware River Treatment facility in Delran, NJ) was attempted to pre-condition problematic pipes using a protocol similar to the NSF protocol for preconditioning pipes (Bellen et al., 1993). Water was periodically exchanged in the pipe segments over the 28-day interval to try to stabilize the pH reactive surfaces. Following this procedure, the pipes were re-exposed to the synthetic buffer water for a total of 7 additional days (Table 5), however, negligible improvements in buffering capacity were recorded in the pipes, so the project team decided to continue with the washed pipes and current synthetic water, while monitoring and recording the pH of future experiments. For cement-lined pipes the pH changes were probably due to dissolution of calcium and silicate oxides from the cement surface that was not stabilized within the test period. Corrosion of the new iron and galvanized surfaces released hydroxide ions through the reaction of iron and the surrounding water.

Table 5. Preconditioning problematic pipes per NSF guidelines.*

Substrate	Day 0 pH	Day 7 pH
Iron	7.2	9.4
New galvanized	7.2	9.2
Cement lined ductile iron (w/o seal coat)	7.2	11.3
Cement lined ductile iron (with seal coat)	7.2	9.3

* Tap water used during 28 day exposure period, changing tap water throughout preconditioning treatment. Synthetic water contained 100/20 mg/L of bicarbonate and carbonate.

Because of the low recovery efficiencies for some inorganic contaminants, particularly on the heavily tuberculated galvanized pipe and cement surfaces, the impact of an extended wash with 0.1M NH₄Cl was evaluated for contact times of <1 min and 24 hours. There was a negligible difference between the two washes for most pipe surfaces (illustrated by the arsenic data in Table 6). The exceptions were cesium and strontium, which had significant increases in recovery during the 24 hr contact wash for the iron-biofilm, used galvanized, and cement lined ductile iron (w/o seal coat) pipe. Strontium also had a significant recovery increase in new iron and galvanized pipes. Additional control testing of un-inoculated pipes was performed to verify that these compounds were not coming from the pipe materials. These experiments showed insignificant background levels of cesium (0.013 mg), but low levels of strontium (1.03 mg)

from the unlined cement surface. For the decontamination experiments, the getter wash with 0.1M NH₄Cl was extended for 24 h (data for the water matrix and the pipe surface experiments utilized the shorter contact time).

Table 6. Comparison of 0.1M NH₄Cl getter wash at <1min and 24 hour contact time

Substrate*	Arsenic mg/L		Cesium mg/L		Strontium mg/L	
	<1min NH ₄ Cl	24hr NH ₄ Cl	<1min NH ₄ Cl	24hr NH ₄ Cl	<1min NH ₄ Cl	24hr NH ₄ Cl
cPVC	0.006	0.004	0.010	0.012	0.018	0.022
cPVC-biofilm	0.009	0.016	0.010	0.044	0.030	0.036
Iron	0.084	0.303	0.081	0.243	1.265	4.206
Iron-biofilm	0.072	0.070	3.463	13.605	2.566	7.786
New galvanized	0.098	0.083	0.038	0.173	0.713	6.315
Used galvanized	0.053	0.510	9.024	13.110	2.598	7.296
Copper	0.315	0.294	0.003	0.005	0.033	0.059
Cement lined DIP w/o seal	0.254	1.879	1.252	26.215	0.832	13.596
Cement lined DIP w/seal	0.058	0.200	0.067	0.731	0.206	0.437
Polyethylene	0.045	0.106	0.093	0.042	0.055	0.035
Epoxy coated steel	0.231	0.164	0.003	0.007	0.010	0.008

*Used galvanized (with biofilm and heavy tuberculation), cement lined ductile iron (w/o seal coat), cement lined ductile iron (with seal coat), and epoxy coated steel pipe

Experimental Reproducibility

Control pipes performed during the decontamination protocol (Phase 2) essentially replicated the procedures used to screen pipe materials in Phase 1b (with the exception of the extended NH₄Cl getter water). It was possible, therefore to compare recovery efficiencies between these two experiments for replicate experiments (Table 7). In general, the results of replicate experiments fell within the expected variability of $\pm 20\%$ (from average). Iron and galvanized pipes showed the lowest overall recovery efficiency. The reason for lower level is not completely clear, but could be related to the presence of corrosion products interfering with the chemical analyses.

Table 7. Reproducibility of recovery efficiencies for replicate experiments.*

Contaminant	cPVC		Fe		Fe-biofilm		New Galvanized		Used Galvanized [#]		Cement lined [#]		Epoxy [#]	
	Phase 1b	Phase 2	Phase 1b	Phase 2	Phase 1b	Phase 2	Phase 1b	Phase 2	Phase 1b	Phase 2	Phase 1b	Phase 2	Phase 1b	Phase 2
Cesium									120	83	43	60		
Strontium									35	57	60	68		
Cobalt			16	7.9			25	36						
Thallium			80	61			53	49						
Chlordane	75	80							68	106			44	120
p-DCB	13	31							49	45			24	60
<i>Bacillus</i> spores					19	2.0			2.0	4.5				

* Comparisons are for contaminant adsorption to pipe materials (Phase 1b) and control experiments during the examination of decontamination methods (Phase 2). Percent recoveries are based on the total mass balance.

[#] Used galvanized (with biofilm and heavy tuberculation), cement lined ductile iron (w/o seal coat), and epoxy coated steel pipe.

The recovery of chlordane was relatively consistent by pipe material, ranging from 68% for used galvanized pipe to 120% for epoxy coated steel pipe. The recovery efficiency of p-DCB ranged from 13% for cPVC to 60% for epoxy coated steel pipe. The p-DCB recovery efficiencies were influenced by the use of 100% methanol which was necessary to solublize the organic compounds, but the methanol reduced the extraction efficiency of the semi-volatile method (Appendix x). Paradichlorobenzene, a solid that is practically insoluble in water, contributed to the solubility problems.

The recovery efficiency of *Bacillus thuringiensis* spores was very low in used galvanized pipe (at 4.5% recovery). The low recovery could be explained by variations in the measurement of the initial stock titers (which were used to determine recovery efficiency). The initial measurement of the *Bacillus* stock was 4.5×10^5 spores/mL, but when retested in the positive control sample (a sterile test tube stored for 7 days); the average count was 2.85×10^5 spores/mL. It is not clear why the stock solution changed. The recovery efficiency of *Bacillus* spores was also very low in iron-biofilm pipes, at 2.0% recovery. In this case the initial measurement of the *Bacillus* stock of 3.3×10^4 spores/mL was lower than expected. It is possible that clumping of the spores prevented accurate enumeration, but this possibility was not examined.

RESULTS

Effect of the Water Matrix

The primary objective of the water matrix evaluation (Phase 1a) was to determine whether the water matrix had a substantial impact on the adsorption of contaminants to the pipe surface. To streamline this evaluation, the studies were limited to iron and cPVC pipe surfaces. The water matrix parameters examined included temperature, pH, hardness, and TOC.

Effect of Temperature

The effect of temperature was examined at room temperature (approximately 20°C) and at 4°C. Overall, there was not a consistent or substantial impact of temperature on the attachment of microbes (*Bacillus* or MS2), inorganic metals, or organic contaminants to cPVC pipe (Figure 3).

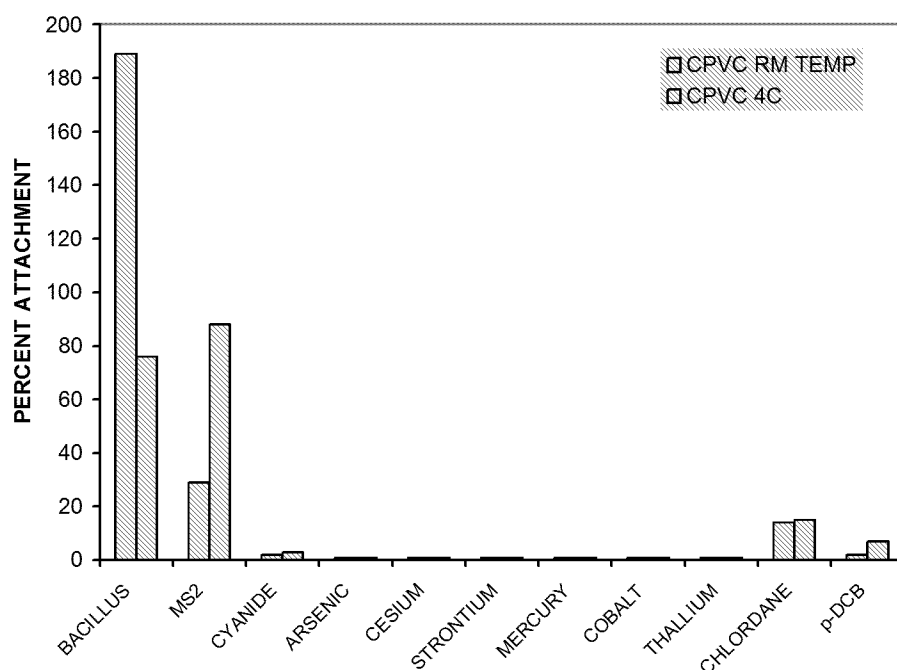


Figure 3. Impact of temperature on the adsorption of contaminants to cPVC pipe.

For iron pipe there was a consistent impact of low temperature on the adsorption of contaminants, but this effect was small – generally less than 5% (Figure 4).

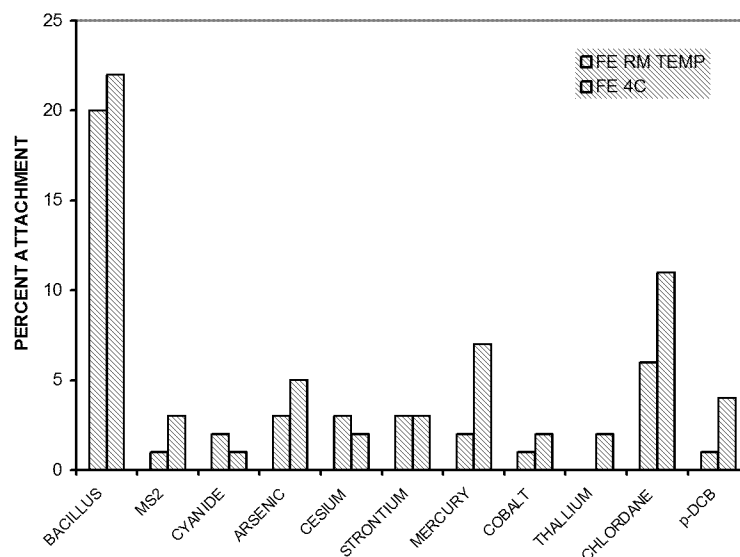


Figure 4. Impact of temperature on the adsorption of contaminants to iron pipe.

Because of the logistical difficulty of conducting all the experiments at cold temperatures and the minor impact of temperature on the adsorption of contaminants to either iron or cPVC surfaces, it was decided to conduct the remainder of the experiments at room temperature.

Effect of pH

A pH of 7.2 or 8.5 did not have a large or consistent effect on the adsorption of organic contaminants on cPVC or iron pipes (Figures 5 and 6). *Bacillus* spores appeared to attach slightly better at pH 7.2 than 8.5 MS2 phages were not examined at pH 8.5 because the phage are not stable at high pH levels.

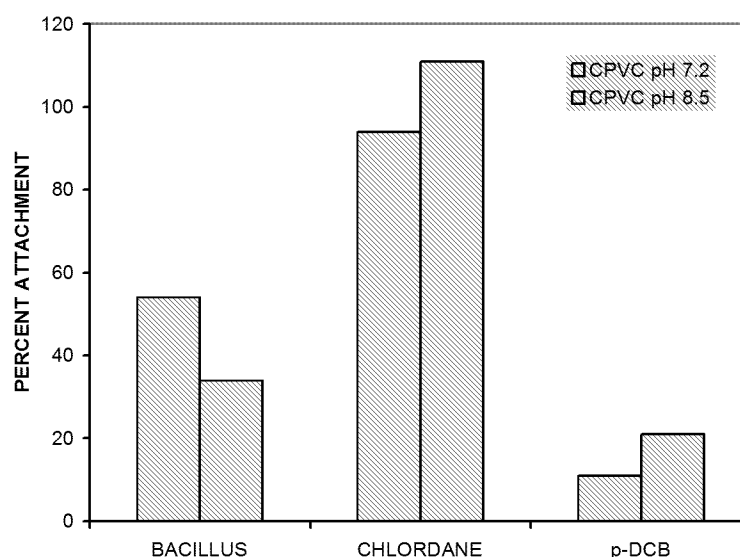


Figure 5. Impact of pH on the adsorption of contaminants to cPVC pipe.

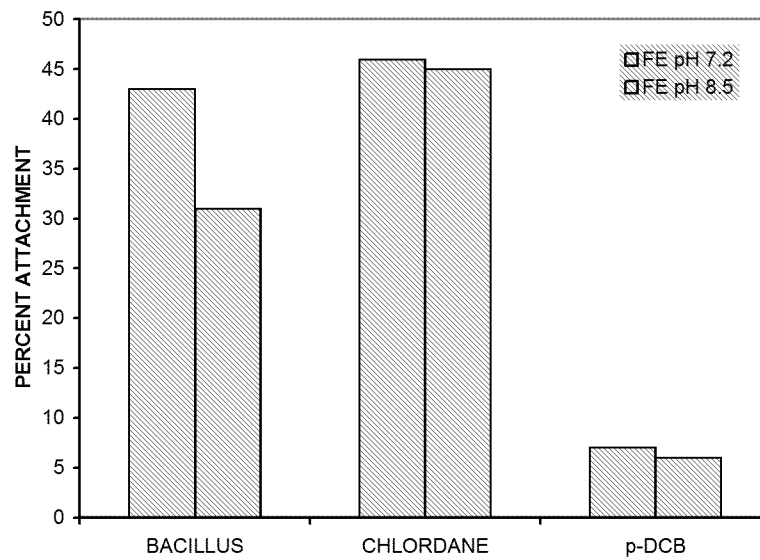


Figure 6. Impact of pH on the adsorption of contaminants to iron pipe.

The adsorption of inorganic contaminants was consistently low (ranging between 0.1 and 1%) and did not have any apparent effect by either pH 7.2 or 8.5.

Because the attachment of *Bacillus* spores was possibly favored at pH 7.2 and because the remainder of the contaminants was not impacted by pH, it was decided to conduct the subsequent experiments at a pH of 7.2.

The water matrix uses a bicarbonate/carbonate and a dihydrogen and monohydrogen phosphate buffer to maintain the pH of the test system. This buffer is similar to natural drinking water and has been successfully used in other experiments. However, in later experiments it was noted that the pH of the water in new iron pipes changed during the incubation period (Table 8). Although the stock solutions were adjusted to a target pH of 7.2, and that this pH was maintained (± 0.5 pH units) in the cPVC pipe system, the pH of the iron pipes rose during the incubation period to over pH 9.0. This effect was evident even after one day incubation and even with varying ratios of bicarbonate and carbonate (Table 8). Corrosion of the iron pipe surface is assumed to cause this effect and may be responsible for the poor recovery efficiencies for some inorganic contaminants.

Table 8. Impact of Pipe materials on the pH stability of the Test System

Water matrix *	pH in cPVC Pipe	pH in Iron Pipe
Initial stock solution	7.20	7.20
1 day incubation 100/20 alkalinity	7.34	9.66
7 day incubation 100/20 alkalinity	6.73	9.50
7 day incubation 100/100 alkalinity	6.84	9.16
7 day incubation 100 alkalinity	7.21	9.48

* The alkalinity ratio refers to the combination of bicarbonate and carbonate added to the synthetic water. All experiments used the same dihydrogen and monohydrogen phosphate concentrations.

The Effect of Alkalinity

The effect of alkalinity was examined using two bicarbonate/carbonate concentrations (100/20 and 100/100 mg/L) representing moderately-hard and hard water. The experimental results did not show any consistent effect of alkalinity on the adsorption of contaminants to cPVC or iron pipe surfaces (Figures 7 and 8). Figure 8 shows adsorption of the inorganic contaminants to iron pipe to be substantially greater at the 100/20 alkalinity. It is thought that these results are an artifact because all other experiments used this same base-line alkalinity and the adsorption of these contaminants to iron pipe were consistently in the 1-5% range.

Because the effect of alkalinity was not apparent, it was decided to use the base-line alkalinity of 100/20 (bicarbonate and carbonate) as this represents moderately hard water and is reflective of typical values seen in many systems.

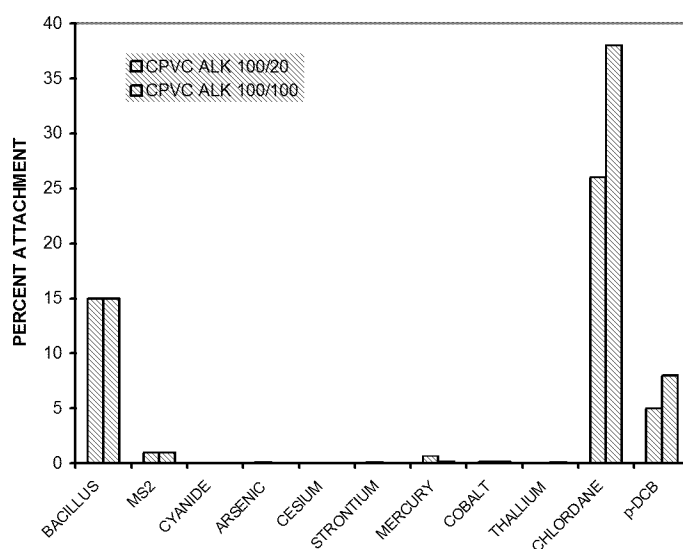


Figure 7. Impact of alkalinity on the adsorption of contaminants to cPVC pipe.

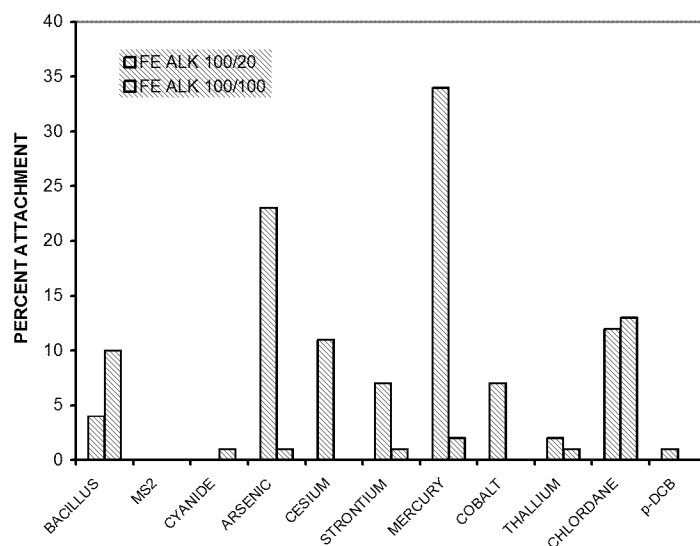


Figure 8. Impact of alkalinity on the adsorption of contaminants to iron pipe.

Impact of TOC

Organic carbon was thought to be potentially important in the interaction between contaminants and the pipe surface. The TOC values of 2 and 4 mg/L were chosen to reflect the typical range of finished drinking waters. Commercially available humic acid was chosen as the source of TOC in these experiments, but other sources of TOC could have different effects. It was beyond the scope of this project to examine various sources of TOC and it was thought that humic acid was a reasonable surrogate of organic matter in natural water.

In these studies a difference between a TOC of 2 or 4 mg/L did not have a dramatic or consistent effect on the adsorption of contaminants to either cPVC or iron pipe surfaces (Figures 9 and 10). For cPVC (Figure 9) there was no overall impact by TOC on contaminant adsorption. For iron pipe surfaces (Figure 10), the adsorption of strontium was very high at a TOC of 4 mg/L and not consistent with results seen in other experiments. For cyanide and arsenic, a TOC of 4 mg/L may be more favorable for adsorption than at 2 mg/L. However, for cobalt and thallium, adsorption values were slightly better at 2 mg/L than at 4 mg/L.

The 4 mg/L humic acid mixture was highly colored and a small precipitate formed after storage for 7 days. Although mixing could resuspend this precipitate, it was decided to use the 2 mg/L TOC solution for subsequent experiments to avoid any complications due to the water matrix formula.

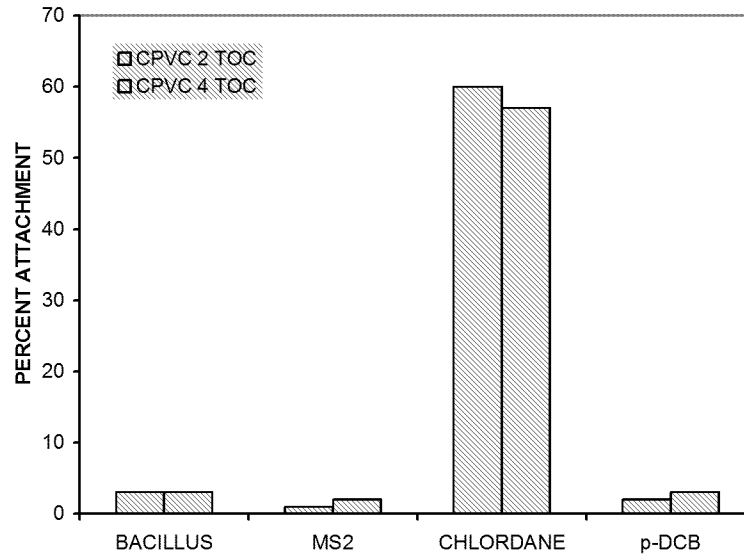


Figure 9. Impact of TOC on the adsorption of contaminants to cPVC pipe.

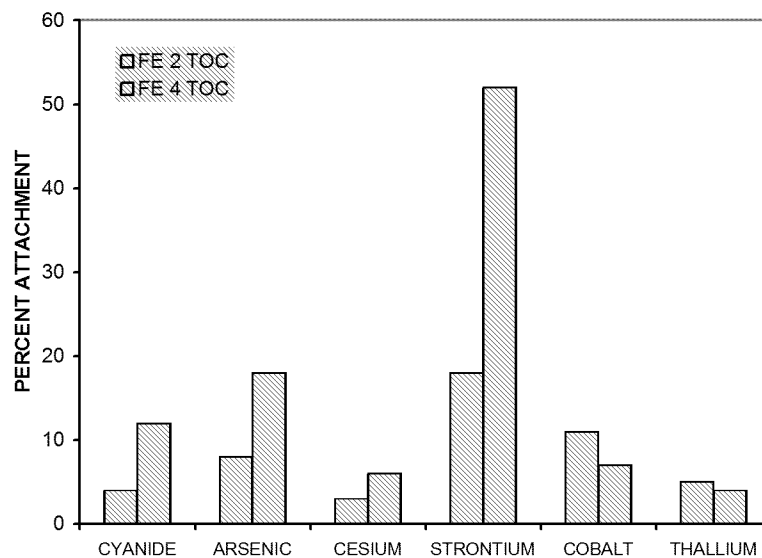


Figure 10. Impact of TOC on the adsorption of contaminants to iron pipe.

Figure 11 summarizes all of the experimental data in a series of Box and Whisker plots. For these plots, the box represents the middle 50 percentile data and the middle line is the mean. The vertical lines, or whiskers, extend from the boxes and represent the upper and lower 1.5 interquartiles. Values falling outside the interquartiles are marked as individual data points.

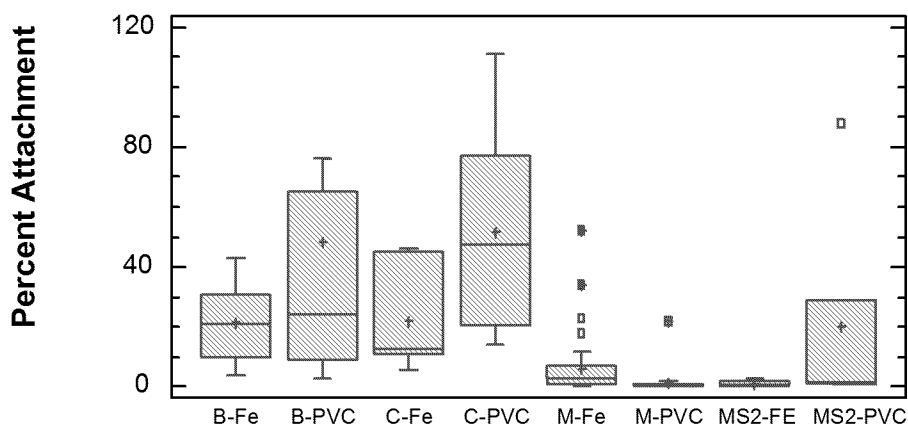


Figure 11. Box and Whisker plot for all contaminant data on iron and cPVC pipe. Abbreviations: B, *Bacillus*; C, chlordanes; M, all inorganic compounds; MS2, MS2 bacteriophage; Fe, iron pipe; PVC, cPVC pipe

Overall the greatest contaminant adsorption was seen with chlordanes and cPVC pipe. Chlordanes attached less well to iron pipe, but values averaged approximately 20%. *Bacillus* spores attached well to both cPVC and iron pipes with median values near 20%. Although *Bacillus* and chlordanes attached at the highest average levels, they also showed the greatest variation (shown by the size of the box). The large variability may suggest that in fact some of the water quality parameters did affect the adsorption of the contaminants to the pipe surfaces, however this difference was not apparent from the limited experiments performed. Additional studies are needed to reduce this variability. The inorganic contaminants were generally easily washed from the pipe surfaces with adsorption rates generally around 1-2%. MS2 bacteriophage had variable attachment to cPVC pipe, but the low values for iron pipe probably were due to the changes in pH and the loss of viability of this phase at high pH.

Change in the Calculation of Attachment

The initial data analysis conducted during the evaluation of the water matrix calculated attachment as the sum of the two washes plus the getter wash. This analysis was subsequently changed to contaminant detected only in the final getter wash eliminate the mass of contaminant held in the water. The change in calculation better reflected the interaction of the contaminant with the pipe surface and facilitated the analysis of the decontamination procedures. Given the low level of adsorption of the inorganic contaminants in the first phase of the research (examination of the water matrix) the change in calculation would not affect the conclusions of the inorganic analyses.

For the organic compounds (chlordan and p-DCB), the majority of the contaminants were obtained in the final getter wash. For chlordan, the average (4 replicates) increase was more than 300 fold over the second wash (Table 9). For p-DCB, the average increase was 5.6 times higher than the previous wash. For *Bacillus* spores the average increase in concentration from the second wash was 0.31 logs or about 2-fold, but for MS2, the final getter wash did not increase phage values in any of the four experiments and the average level was 0.24 logs less than the second wash.

Table 9. Impact of the final getter wash on contaminant desorption from cPVC pipe

Contaminant	N	Wash 1	Wash 2	Getter Wash
<i>Bacillus</i> (log/mL)	4	3.91	2.83	3.14
MS2 (log/mL)	4	3.70	1.86	1.62
Chlordan (mg/L)	4	0.385	0.122	37.680
p-DCB (mg/L)	4	0.723	0.230	1.281

N, number of replicate experiments

These results also support the contention that the change in calculation for adsorption (to only the final getter wash value) would not change the conclusions of the water matrix studies, although the amount of microbial attachment might be somewhat less than reported in Figure 11.

Adsorption of Contaminants to Pipe Surfaces

Data presented in Table 10 show the interaction of the inorganic, organic and microbial contaminants with eleven pipe surfaces. For these results, the contaminant adsorption is reported as the percent ratio of the mass of the final getter wash to the mass of the initial inoculum. An arbitrary value of 5% was chosen to select contaminant and pipe combinations for subsequent decontamination testing. The 5% cut-off ensured that sufficient contaminant levels remained on the surface to evaluate a 100-fold reduction (based on the analytical detection levels) to observe by decontamination procedures for contaminants that adsorb at levels greater than 5% (about 5 mg/L).

The pipe materials included combinations that allowed the evaluation of biofilms on cPVC and iron pipe, clean and tuberculated galvanized pipe surfaces, and cement-lined ductile iron pipe with and without the asphaltic seal coat. For inorganic contaminants, cesium, strontium, cobalt and thallium attached at rates greater than 5% to iron, new galvanized, or iron/used galvanized-biofilm pipes (Table 10). Chlordan adsorbed to all pipe surfaces except the cement lined ductile iron and polyethylene pipes. Although the p-DCB adsorption rates were much lower than those for chlordan, p-DCB attached at rates greater than 5% to iron-biofilm, new galvanized, used galvanized, cement lined ductile iron (with seal coat), and epoxy coated steel pipe. *Bacillus* spores attached best to iron-biofilm pipe.

Table 10. Percent Adsorption[†] of Contaminants to Various Pipe Surfaces*

Contaminant	cPVC	cPVC Biofilm	FE	FE Biofilm	New GALV	Used GALV*	CU	Cement*	Cement w/Seal*	PE	Epoxy*
CN	0.0	0.0	0.7	0.9	2.6	0.0	0.0	1.2	0.0	0.0	0.3
As	0.0	0.0	0.3	0.3	0.4	0.2	1.4	0.9	0.2	0.2	1.0
Cs	0.0	0.0	0.1	4.6	0.0	12.0	0.0	1.3	0.1	0.1	0.0
Sr	0.1	0.1	3.5	8.0	2.0	8.1	0.1	2.3	0.6	0.2	0.0
Hg	0.4	2.6	1.2	0.8	0.2	0.7	1.7	0.5	0.5	4.9	0.7
Co	0.1	0.2	0.6	1.4	6.7	3.4	0.2	0.1	3.7	1.4	0.2
Tl	0.0	0.0	6.6	1.8	3.1	7.2	0.0	0.8	0.2	0.4	0.0
Chlordane	45.8	32.4	15.7	23.2	28.9	27.7	18.3	1.5	2.3	1.5	18.3
p-DCB	1.3	0.9	1.1	5.4	9.1	5.7	0.1	2.7	9.3	0.4	16.2
Bacillus	0.0	0.0	2.0	27.0	1.0	3.0	0.0	0.0	2.0	0.0	1.0
MS2	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0

[†] Measured as the mass of the getter wash divided by the mass of the inoculated stock solution.

* Used galvanized (with biofilm and heavy tuberculation), cement lined ductile iron (w/o seal coat), cement lined ductile iron (with seal coat), and epoxy coated steel pipe

Biofilms

Similar pipe materials with and without biofilms provided an opportunity to compare the impact of clean and colonized surfaces on the adsorption of various contaminants. Paired samples of cPVC, black iron, and galvanized pipe were used for these comparisons. To cultivate the biofilms, pipes were filled with synthetic water and incubated at room temperature for 14 days. To determine the biofilm levels, the synthetic water was drained off and the pH of the buffered water analyzed. Pipe surfaces were scraped using the same procedure as the microbial “getter wash.” Heterotrophic Plate Counts (HPC’s) were performed in duplicate using R₂A agar and quantified as cfu/cm². HPC levels generally ranged between 10⁴ and 10⁵ cfu/cm². Biofilm levels were checked periodically throughout the experimental phase for variability but all subsequent biofilm samples showed good agreement between replicates (between 10⁴ and 10⁵ cfu/cm²).

Consistent with previous results (Tables 3 and 10), biofilms appeared to improve the adsorption of some contaminants while not affecting others. Cesium and strontium appeared to increase adsorption to iron and used galvanized pipe with biofilms, while biofilms on cPVC had little effect. It is possible that biofilm bacteria on iron or galvanized pipe produced reduced iron species which could complex these metals. Biofilms on cPVC would have no such effect.

Bacillus spores appeared to attach to iron pipes with biofilms better than clean pipes alone. Previously published studies have shown that *Cryptosporidium* oocysts and some viruses are entrapped and accumulated in biofilm samples (Quignon, et al. 1997; Piriou et al., 2000).

Understanding the role of biofilms in contaminant adsorption is important because it may be possible to formulate decontamination strategies to remove biofilms in addition to the specific contaminants. For example, it was of interest to use high chlorine doses for decontamination of cesium or strontium since these contaminants seem to have increased adsorption to biofilms.

Effectiveness of Extended Washes - Persistence of Adsorbed Contaminants

Limited experiments were performed with the inorganic contaminants to examine the impact of multiple washes to characterize the nature of the interaction of the contaminants with the pipe surface. In these studies the pipe segments were washed a total of 8 times (7 washes and a final ammonium chloride getter wash). Experiments were performed for both iron and cPVC pipe, but only the data for the cPVC pipe are shown (Figure 12) because the results were representative for both surfaces. Data shown in Figure 12 indicate that the initial two washes reduce the concentration of the contaminants by 1,000-fold, but subsequent washes (up to six washes) result in only an additional 10-fold reduction in concentration. The final “getter wash” did not result in a substantial increase in concentration, suggesting that the metals are simply being diluted from the pipe surface with diminishing efficacy.

The results were similar to the full-scale results of the chlordane contamination incident (see Appendix A) where initial flushing of the distribution system resulted in rapid decreases in chlordane concentrations, but trace levels of chlordane persisted in the system for an additional 6 months before finally being eliminated. These results are important for shaping a utility’s communications and expectations following a contamination incident. Incident managers should be prepared to communicate the expectation that low levels of contaminants may be detected for prolonged periods, even after initial decontamination protocols have significantly reduced the initial concentrations.

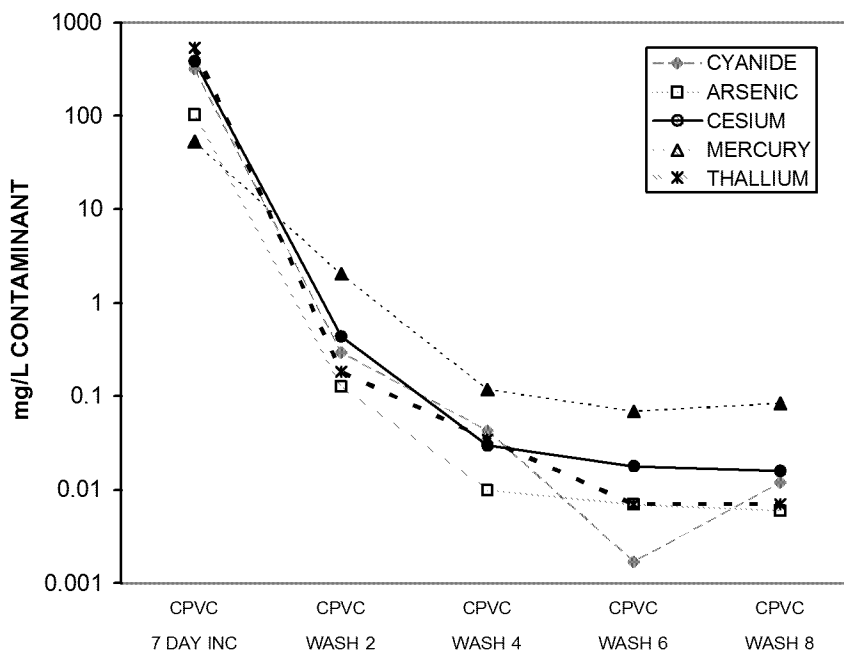


Figure 12. Impact of multiple washes on the desorption of inorganic contaminants from cPVC pipe.

Impact of Contact Time

It was of interest to examine the impact of contact time on the exposure of contaminants to pipe surfaces to determine the importance of response times for infrastructure decontamination. Exposure times of 1 hour, 24 hours, and 7 days for chlordane and p-DCB added to cPVC pipes showed that the longer exposure times increased the attachment of both contaminants (Figure 13). Therefore, quick response times for infrastructure decontamination are critical when dealing with organic contaminants.

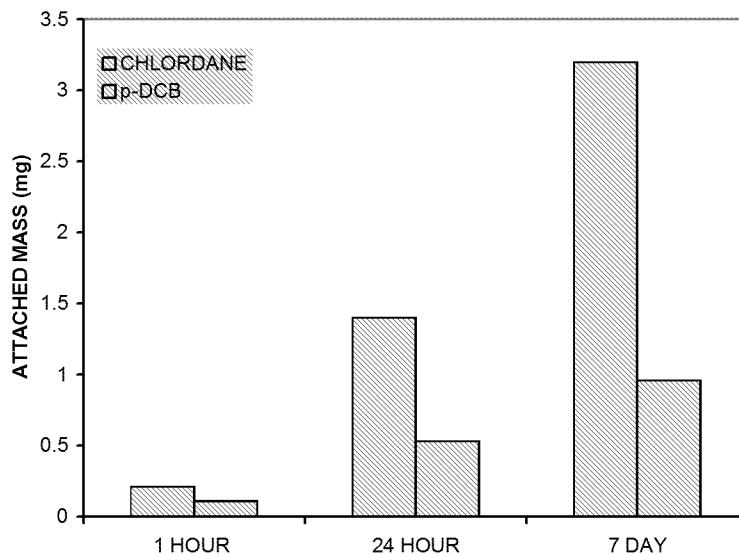


Figure 13. Organic mass attachment on cPVC pipe over time.

Decontamination of Pipe Surfaces

The effectiveness of the decontamination procedures was evaluated by comparing the ability of the decontamination agents to reduce contaminants levels below those observed by the water wash (control) alone (Figure 2). Based on the analytical detection levels, the experiments had the ability to observe a 100-fold reduction by the decontamination procedures. The test conditions were selected for contaminants that adsorbed to pipe surfaces at levels greater than 5% (Table 10).

Bacterial Decontamination

Results for *Bacillus* spores attached to heavily tuberculated galvanized pipe assay indicated that various chlorine contact times (expressed as target CT's, the chlorine concentration

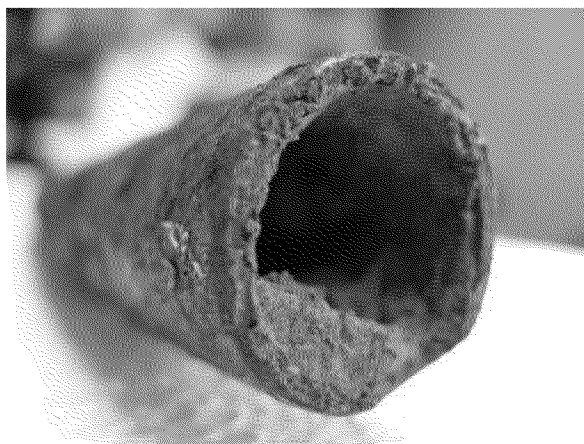


Figure 14. Heavily tuberculated used galvanized pipe.

[mg/L] multiplied by the contact time [minutes]) did not dramatically inactivate the spores. *Bacillus* spore inactivation ranged from 43 to 84 percent (Table 11). Because of the extreme level of corrosion tubercles on the used galvanized pipes (Figure 14), the chlorine demand was excessive and chlorine residuals could not be maintained during the assays. Chlorine plus a physical cleaning procedure, such as pigging, will be needed to achieve complete removal of spores on such pipes, and decontaminating these pipes may be a challenge in real world situations. Results from the *Bacillus thuringiensis* on iron-biofilm pipe assay also showed excessive chlorine demand. Chlorination removed the attached *Bacillus*; however, the overall effectiveness is questionable since the level of attached spores was very low (Table 12).

Table 11. Percent inactivation of *Bacillus thuringiensis* by chlorine on used galvanized pipe.

Target Chlorine CT*	Chlorine Dose (mg/L)	Contact Time (min)	Residual Chlorine (mg/L)	Attached Spore Count	Percent Removal
Control	-	-	-	69,000	-
300 mg/L-min	50	6	9	25,000	65
3,000 mg/L-min	50	60	1.6	40,000	43
30,000 mg/L-min	25	1200	<0.02	11,000	84

*All experiments performed at room temperature with baseline parameters of pH 7.2, 2.0mg/L TOC, 100/20 alkalinity ratio. Target chlorine CT levels were not achieved due to excessive chlorine demand.

Table 12. Percent inactivation of *Bacillus thuringiensis* by chlorine on iron-biofilm pipe.

Target Chlorine CT*	Chlorine Dose (mg/L)	Contact Time (min)	Residual Chlorine (mg/L)	Attached Spore Count	Percent Removal
Control	-	-	-	500	-
300 mg/L-min	50	6	16.6	0	100
3,000 mg/L-min	50	60	0.14	0	100
30,000 mg/L-min	100	300	<0.02	250	50

*All experiments performed at room temperature with baseline parameters of pH 7.2, 2.0mg/L TOC, 100/20 alkalinity ratio. Target chlorine CT levels were not achieved due to excessive chlorine demand.

Isolates surviving decontamination were identified by a commercial laboratory (Microbial ID, Newark, DE) by DNA sequencing and showed that both the spiked *Bacillus thuringiensis* and naturally occurring *Bacillus cereus* were present. Examination of tubercles on a non-exposed galvanized pipe showed that *Bacillus* spores including *Bacillus thuringiensis*, *Bacillus megaterium*, and *Bacillus pumilus* occurred naturally in the pipes. The presence of *Bacillus thuringiensis* spores in natural biofilm samples opens the possibility that some of the surviving spores may have originated from the pipe tubercles and not from the added inoculum.

A commercially available pipe cleaning chemical (RE-Ox) was used with a different strategy; rather than removing the contaminant the strategy was to remove the tuberculated deposits. RE-Ox is normally applied for a number of weeks in a flowing system (<http://www.re-ox.com/>), so the static 24 hour exposure used in the decontamination protocol was not the usual application of the product. Even though the product is measured using free chlorine, the product is not a

licensed biocide. Exposure of a spore suspension to the diluted (100x) product (measured chlorine residual of 6 mg/L) for 24 hours did not result in any decrease in spore viability. Exposure of the tuberculated pipe to the concentrated RE-Ox product for 24 hours resulted in a small decrease in the level of pipe tuberculation (Figure 15). *Bacillus thuringiensis* spore counts did not decrease during this exposure (Table 13). Although pipe cleaning may be an effective decontamination approach when applied in full-scale systems, it was not effective under the constraints of the protocol tested in this study.

Table 13. Percent inactivation of *Bacillus thuringiensis* using RE-Ox on used galvanized pipe.

Target RE-Ox CT*	RE-Ox Dose (mg/L)	Contact Time (min)	Residual Chlorine (mg/L)	Attached Spore Count	Percent Removal
Control	-	-	-	51,000	-
756,000 mg/L-min	525	1440	0.11	52,500	-2.9
79,200 mg/L-min	55	1440	<0.02	75,300	-48
7,200 mg/L-min	5	1440	<0.02	107,400	-110

*All experiments performed at room temperature with baseline parameters of pH 7.2, 2.0mg/L TOC, 100/20 alkalinity ratio. Residual chlorine was measured at the end of the contact time.

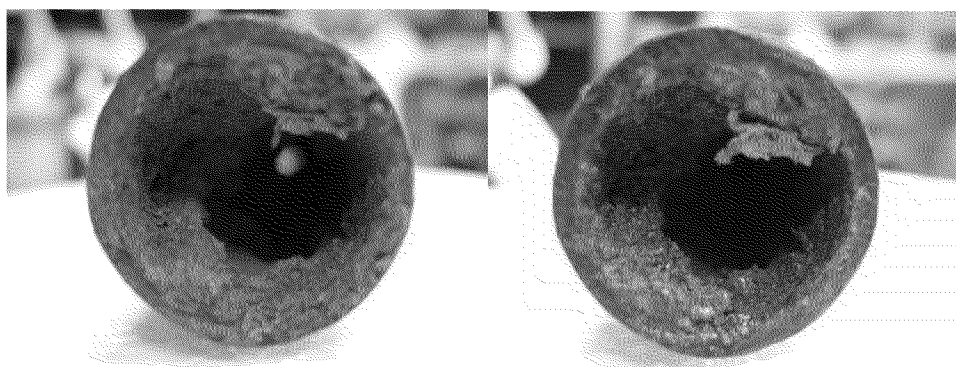


Figure 15. Heavily tuberculated used galvanized pipe before (left) and after (right) RE-Ox treatment.

Decontamination of Inorganic Contaminants

Two pipe surfaces (heavily tuberculated galvanized pipe, and cement lined ductile iron pipe without seal coat) were contaminated with inorganic compounds and then treated with chlorine and two concentrations of Simple Green (a commercially available detergent and chelator) with minimal success. Chlorine showed some enhanced removal of cesium on used galvanized and cement lined ductile iron (without seal coat) at 23 and 26 percent, respectively (Table 14). Strontium was also minimally removed on cement lined DIP (without seal coat) by chlorine. The 10% Simple Green aided in the removal of cesium and strontium with efficiencies ranging between 18 and 56 percent (Table 14). However, none of the decontamination agents worked successfully on cobalt or thallium attached to new iron or galvanized pipe (Table 15). The negative values for the removal of the metals by decontamination agents indicate that the synthetic water washes were more effective at removing cobalt and thallium on both iron and

galvanized pipe. The negative values could also be related to the relatively low level of adsorption of the metals to these surfaces and small variations in analytical measurements. Treatment of cement lined ductile iron pipe (w/o seal coat) using EDTA and sodium citrate showed that neither was effective for removing cesium. However, 1% sodium citrate was minimally effective (34%) for removal of strontium (Table 14).

Table 14. Percent cesium and strontium removal by various decontamination agents.

Decontamination Agent	Pipe Substrate*	Cesium Attached Mass (mg)	Cesium Percent Removal	Strontium Attached Mass (mg)	Strontium Percent Removal
Control	Used Galvanized	0.990	-	0.670	-
30,000 mg/L-min Cl ₂ [^]	Used Galvanized	0.763	23	0.722	-7.8
1% Simple Green	Used Galvanized	1.20	-21	0.829	-24
10% Simple Green	Used Galvanized	0.525	47	0.298	56
Control	Cement line DIP	34.1	-	14.2	-
30,000 mg/L-min Cl ₂ [^]	Cement line DIP	25.3	26	9.28	35
1% Simple Green	Cement line DIP	21.8	36	6.70	53
10% Simple Green	Cement line DIP	18.8	45	6.20	56
Control	Cement line DIP	40.0	-	20.3	-
1% Na EDTA	Cement line DIP	46.1	-15	19.0	6.3
1% Na Citrate	Cement line DIP	45.5	-14	13.4	34
10% Simple Green	Cement line DIP	32.8	18	15.1	26

[^] Chlorine dosed at 25mg/L for 20 hours to achieve targeted chlorine contact time.

* Used galvanized (with biofilm and heavy tuberculation) and cement lined ductile iron pipe (w/o seal coat).

Table 15. Percent cobalt and thallium removal by various decontamination agents.

Decontamination Agent	Pipe Substrate	Cobalt Attached Mass (mg)	Cobalt Percent Removal	Thallium Attached Mass (mg)	Thallium Percent Removal
Control	Iron	0.035	-	1.187	-
30,000 mg/L-min Cl ₂ [^]	Iron	0.055	-58	1.568	-32
1% Simple Green	Iron	0.132	-280	0.354	70
10% Simple Green	Iron	0.074	-110	1.108	6.7
Control	New Galvanized	0.045	-	0.030	-
30,000 mg/L-min Cl ₂ [^]	New Galvanized	0.165	-270	0.056	-87
1% Simple Green	New Galvanized	0.091	-100	0.082	-170
10% Simple Green	New Galvanized	0.113	-150	0.235	-680

[^] Chlorine dosed at 25mg/L for 20 hours to achieve targeted chlorine contact time.

Decontamination of Organic Contaminants

Three surfactants (N-60, TDA-6, and LZV) were applied to cPVC pipe to determine their effectiveness for removal of chlordane and p-DCB. N-60 and TDA-6 showed significant removals of chlordane attached to cPVC pipe (Table 16) with increasing removals with increasing concentrations of surfactant. The LZV surfactant did not show significant removal of

Table 16. Percent chlordane and p-DCB removal for various detergents from cPVC pipe.

Decontamination Agent	Attached Mass (mg) Chlordane	Chlordane Percent Removal	Attached Mass (mg) p-DCB	p-DCB Percent Removal
Control	3.23	-	0.962	-
0.05% N-60	1.44	55	1.01	-5.4
0.5% N-60	0.696	79	1.01	-5.4
5% N-60	0.397	88	0.826	14
Control	3.23	-	0.962	-
0.05% TDA-6	1.61	50	1.03	-6.8
0.5% TDA-6	0.657	80	0.943	2.0
5% TDA-6	0.390	88	0.832	14
Control	3.23	-	0.962	-
0.05% LZV	3.23	0.0	1.05	-9.5
0.5% LZV	1.49	54	1.04	-8.1
5% LZV	1.63	50	0.852	11

Abbreviations: Surfonic N-60, nonylphenol ethoxylate; Surfonic TDA-6, ethoxylated tridecyl alcohol; Empicol LZV, C12-C18 sulfate sodium salt.

chlordane at 0.05% concentration, but was equally effective using either the 0.5% or 5% concentration. The effects on p-DCB decontamination were quite different, and N-60, TDA-6 or LZV had virtually no effectiveness for removing p-DCB from cPVC pipe. In fact, none of the surfactants performed significantly better at p-DCB removal than using the synthetic water wash alone. Again the low removal efficiency could be related to the relatively low adsorption of p-DCB to the pipe surface.

For chlordane and p-DCB applied to used galvanized pipe, the N-60 and TDA-6 surfactants showed good removals of chlordane with both showing increased contaminant removal with increasing concentrations (Table 17). LZV did not show effective removal of chlordane at the 0.5% concentration, but was effective at the 0.05% and 5% concentrations. The effects of the surfactants for p-DCB were somewhat mixed with N-60 showing some effectiveness at the 0.5% and 5% concentration, while TDA-6 was effective at 0.05% and 5%. LZV was minimally effective at removing p-DCB at any concentration.

Table 17. Percent chlordane and p-DCB removal for various detergents from used galvanized pipe.

Decontamination Agent	Attached Mass (mg) Chlordane	Chlordane Percent Removal	Attached Mass (mg) p-DCB	p-DCB Percent Removal
Control	3.13	-	0.245	-
0.05% N-60	2.26	28	0.231	5.7
0.5% N-60	1.18	62	0.143	42
5% N-60	0.622	80	0.198	19
Control	3.13	-	0.245	-
0.05% TDA-6	2.60	17	0.114	53
0.5% TDA-6	1.10	65	0.192	22
5% TDA-6	0.342	89	0.078	68
Control	3.13	-	0.245	-
0.05% LZV	1.53	51	0.267	-8.8
0.5% LZV	3.62	-16	0.208	15
5% LZV	0.351	89	0.208	15

Abbreviations: Surfonic N-60, nonylphenol ethoxylate; Surfonic TDA-6, ethoxylated tridecyl alcohol; Empicol LZV, C12-C18 sulfate sodium salt.

For chlordane and p-DCB applied to epoxy coated steel pipe, all three surfactants showed significant removals of chlordane (Table 18). The surfactants showed increasing effectiveness for contaminant removal with increasing concentrations. The LZV surfactant did not show effective removal of chlordane at the lowest concentration (0.05%), but it was effective at the higher levels. The effects of the surfactants on p-DCB decontamination of epoxy coated steel were very poor. None of the surfactants aided in removal of p-DCB on epoxy coated steel pipe over water wash alone. In contrast to the other pipe materials, p-DCB attached to the epoxy coated steel pipe at levels ranging between 5.6 and 8.5 mg/L (Table 18).

Table 18. Percent chlordane and p-DCB removal for various detergents from epoxy coated steel pipe.

Decontamination Agent	Attached Mass (mg) Chlordane	Chlordane Percent Removal	Attached Mass (mg) p-DCB	p-DCB Percent Removal
Control	13.5	-	6.58	-
0.05% N-60	11.1	18	5.63	14
0.5% N-60	8.93	34	8.48	-29
5% N-60	6.50	52	8.48	-29
Control	13.5	-	6.58	-
0.05% TDA-6	9.19	32	6.20	5.7
0.5% TDA-6	7.74	43	8.46	-29
5% TDA-6	5.32	61	6.59	-0.20
Control	13.5	-	6.58	-
0.05% LZV	15.6	-16	7.75	-18
0.5% LZV	6.27	54	7.72	-17
5% LZV	3.51	74	7.29	-11

Abbreviations: Surfonic N-60, nonylphenol ethoxylate; Surfonic TDA-6, ethoxylated tridecyl alcohol; Empicol LZV, C12-C18 sulfate sodium salt.

DISCUSSION

Protocol Development

One objective of this project was to develop a protocol for testing the attachment of contaminants to various pipe surfaces and to evaluate procedures for decontaminating the surfaces. The protocol selected used short pipe sections to facilitate the testing and handling of the materials. As opposed to a flowing pipe loop system, the pipe section protocol could be used in an emergency to relatively quickly screen cleaning options, especially if contaminant analysis was available on-site.

Impact of Water Matrix

The water matrix testing summarized the development of pipeline testing protocols for the adsorption of biological, inorganic and organic compounds to common pipe surfaces. It examined the impact of water matrix parameters including temperature, pH, hardness, and TOC on the attachment of contaminants to cPVC and iron pipe surfaces. The purpose of these studies was to develop a standardized protocol that can be used to examine a variety of pipe surfaces in the adherence to pipe substrates phase. Therefore, for this portion of the work, the focus was to understand how the characteristics of the water matrix might influence the adsorption of contaminants.

It was a surprise that none of the parameters examined had a consistent or dramatic impact on the attachment of contaminants to either PVC or iron pipes. Although each experiment was performed only once, the level of attachment was considered for five different sets of contaminants (1, *Bacillus* and MS2 microbes; 2, cyanide; 3, arsenate, strontium, and cesium; 4, cobalt and thallium; and 5, chlordane and p-DCB). Therefore, if a water matrix variable such as low temperature (4°C) had a dramatic impact on the attachment of contaminants to pipe surfaces, there would have been ample experimentation to discern a trend. This is not to say that specific conditions may be important for select contaminants, but only that there were not uniform conditions important for all contaminants. Temperature was thought important because some organic contaminants would become more viscous at low temperatures, thus making it more difficult to wash off the pipe surface. Chlordane, however, attached well to the pipes even at 20°C, with levels ranging from 15 to 46 percent (Table 10).

The pH of the test water was also initially thought to be important in attachment as it could affect the solubility of especially inorganic contaminants. The pH levels examined (7.2 and 8.5) represent typical ranges found in drinking water supplies. In some cases we were unable to control the pH of the experiment due to reactions with the surfaces of the new pipes, despite 30 days of preconditioning with tap water (Table 5). Data from the initial testing of the water matrix (figures 5 and 6) was confirmed during the pipe material evaluations (Table 10) in showing that pH had little impact on contaminant attachment. For example, used galvanized pipe was able to maintain a stable pH between 7.35 and 7.51, but fresh galvanized pipe increased the pH level to >9.0 (Table 4). It would be expected that solubility levels of inorganic contaminants would decrease at higher pH levels, but data in Table 5 showed that the level of attachment was not different for most inorganic contaminants and was higher for cesium and

strontium for the used galvanized pipe at pH levels near 7.2. The highest pH levels were experienced on the cement lined ductile iron pipe, with or without a seal coat (pH levels ranging between 9.3 and 11.4), but attachment of inorganic contaminants to these surfaces was generally low (0 to 3.7 %).

Changes in the total organic carbon (TOC) levels were examined because organic complexes could both enhance or inhibit the binding of contaminants to pipe surfaces. Organic carbon could competitively block binding sites on pipe surfaces for microbes and organic contaminants, or organic-metal complexes could improve the adsorption of these contaminants to the pipe surface. For this study, TOC levels were varied between 2 and 4 mg/L using a commercial humic acid. It is likely that other organic carbon compounds would have varying effects, and these should be examined in future experiments. Again, by selecting new and used pipe the impact of adsorbed natural organic compounds can be examined. The used galvanized pipe was taken from an active distribution system that was exposed to a mixture of natural organic matter. Alternatively the new galvanized pipe was exposed only to the 2 mg/L of humic matter that was contained in the test water matrix. However, chlordane attachment to both pipes was nearly identical at 28.9% and 27.7% for new and used galvanized pipe, respectively (Table 10). Attachment of *p*-DCB was slightly higher for new rather than used galvanized pipe (9.1% and 5.7%, respectively), but *Bacillus* spore attachment was slightly better for the used galvanized pipe (3.0% for used pipe compared to 1.0% for new galvanized pipe). Therefore, the overall conclusion is that there is no obvious effect of natural organic matter on contaminant inhibition or attachment, but that the evidence for this conclusion is limited and should be examined in more detail.

Impact of Pipe Surface

The objective of the adherence to pipe substrate phase was to examine the association of contaminants with a variety of pipe surfaces. Two of the pipe selected (cPVC and black iron) were also used in water matrix testing and were used as controls to compare the data from the first two project phases. Heavily corroded used galvanized service line pipe was obtained from a participating water utility, while clean galvanized pipe was used to compare the impact of tuberculation. Biofilms were grown on cPVC, iron, and the tuberculated used galvanized pipes by incubating the pipes with the synthetic water (containing 2 mg/L TOC) for 2 weeks.

Overall, the least attachment of contaminants was observed with medium density polyethylene pipe (Table 10). Even chlordane, which attached at a rate of 46% to PVC, showed little attachment to polyethylene pipe. Polyethylene is known to be resistant to permeation and reactions with chemical contaminants, making it a preferred lining material for hazardous waste sites (Koerner 1998). The typical composition of polyethylene pipe is nearly 97% resin with small amounts of pigment and additives, in contrast to PVC pipe which typically has 90% resin, and 10% stabilizers, pigments and other additives (Koerner 1998). The presence of these additives provides opportunities for a variety of interactions with the chemical contaminants. Chlordane, for example, was dissolved in a kerosene solution which could attack and permeate the PVC pipe (Thompson and Jenkins 1987). Chlordane itself, however, has been shown not to permeate PVC pipe (Lee and Kelleher 1984), however, other pesticides including Lindane, 2,4-D, Banvel-D, and Dinoseb permeated PVC pipes within periods of 1 to 3 weeks (Thompson and

Jenkins 1987). It may be necessary to replace the distribution system pipes should contaminant materials permeate and weaken the mechanical strength of the system. Experiments showed that increasing the contact time with the pipe material increased the amount of adsorbed chlordane and p-DCB (Figure 13). Prolonging the contact time could also increase the level of pipeline permeation for pipe materials and contaminants that are susceptible to permeation. For many compounds, permeation can occur within 1-8 days (Lee and Kelleher 1984). It is important, therefore, that response times for infrastructure decontamination be rapid when dealing with organic contaminants, especially in systems with extensive amounts of PVC pipes.

Cement-lined ductile iron pipe initially showed little interaction with chemical and microbial contaminants, but when longer contact times were used for the 0.1 M NH_4Cl getter wash (Table 6), the data showed increased recoveries of cesium and strontium. Control experiments showed insignificant background levels of cesium (<0.04% of the spiked control), but low levels of strontium (7.3% of the spiked control) from the unlined cement surface. The high pH levels associated with the cement lining resulted in inactivation of the MS2 bacteriophage. It is not clear, however, whether aged cement-lined pipes would have a similar antimicrobial pH effect. The asphaltic seal-coat on the treated cement-lined pipe showed some interaction with p-DCB, resulting in 9.3% attachment, whereas the uncoated cement pipe showed only a 2.7% attachment for p-DCB (Table 10). These results demonstrate that the interaction of various contaminants and pipe surfaces can be highly specific and that coating or modifications to the surface can greatly affect contaminant binding.

Kirmeyer et al. (1994) reported that of the 880,000 miles (1,416,200 km) of mains in service in the United States (1994 data), 57.9 percent had cement linings or were composed of cement. The survey did not distinguish whether these surfaces had an asphaltic seal-coat or not. Approximately 24.6 percent of pipe was unlined cast iron or ductile iron pipe, and 9.2 percent was PVC. The researchers reported that 13,200 miles of new pipes (21,240 km) are installed each year, predominately comprised of cement-lined ductile iron (47.7 percent), PVC (38.7 percent) and concrete pressure (12.5 percent) pipe. Therefore, the majority of distribution mains in the US are cement lined and relatively low susceptibility to interaction with chemical or microbial contaminants, but a growing inventory PVC pipe could be susceptible to contamination by organic compounds.

The presence of biofilms and corrosion tubercles were found to have significant effects on the attachment of contaminants to the pipe surface. Similar pipe materials with and without biofilms provided an opportunity to compare the impact of clean and colonized surfaces on the adsorption of various contaminants. Cesium and strontium appeared to increase adsorption to iron and used galvanized pipe with biofilms. In addition, *Bacillus* spores appeared to attach to iron pipes with biofilms better than clean pipes alone. Previously published studies have shown that *Cryptosporidium* oocysts and some viruses are entrapped and accumulated in biofilm samples (Quignon, et al. 1997; Piriou et al., 2000). The increased surface roughness caused by corrosion tubercles would both increase contaminant binding due to increased surface area, and the cracks and crevices of the tubercles could entrap contaminants. In addition, the various environments within the biofilm and at the surface of the pipe material would cause both oxidizing and reducing conditions that would enhance contaminant interactions through adsorption, precipitation, and chemical reactions with microbial extracellular polymer substances, natural

organic matter, reduced inorganic compounds (e.g., Fe^{+2} , NH_3 , and S^{-2}), and adsorbed particles (Characklis and Marshall 1990).

Pipeline Decontamination

The objective of the examination of decontamination methods was to examine various decontamination strategies based on contaminant chemistry, pipe substrate, and biofilm presence. The successful strategies were evaluated based on the effectiveness of the decontamination agents for contaminant removal.

Chlorine was chosen based on its availability and effectiveness for disinfection of distribution systems. A 3 log inactivation (99.9%) of the *Bacillus thuringiensis* was estimated based on published values for anthrax inactivation (Table 1). CT (the concentration of disinfectant multiplied by the contact time) values of 300, 3000, and 30,000 mg·min/L were targeted to provide disinfection levels roughly 1, 10, and 100 times that necessary for inactivation of suspended spores. AWWA standards for decontamination of various infrastructures (C651-99, C652-02, C653-03, C654-97) all provide for high chlorine doses (10 to 100 mg/L) for extended periods of time (3 to 24 hours). All of these protocols, in theory, provide adequate CT levels for at least 99.9 percent spore inactivation. However, this study demonstrated that highly corroded surfaces could exert sufficient chlorine demand to protect *Bacillus* spores even after high chlorine doses for extended contact times. Of course, the AWWA standards require maintenance of a specified free chlorine residual, which would not have been met in the corroded pipe experiments. In this case, the pipes would need to be cleaned of the tuberculation prior to disinfection. Provided the pipes were in reasonably good shape, it is expected that super-chlorination should be an effective treatment process for nearly all microbial contaminants, except *Cryptosporidium*. For *Cryptosporidium*, high levels of chlorine dioxide could be used for inactivation of oocysts (Arora et al. 2001).

Strategies to remove biofilm and scale could be used to remove attached contaminants. In this study bacterial biofilms were scraped using a test-tube brush as a surrogate for foam pig. Previous research has shown this method to be effective for quantitative removal of attached bacteria (LeChevallier et al. 1990, 1993). A commercially-available pipe cleaning process (Re-Ox) was examined as either a disinfectant or for biofilm removal. Unfortunately the commercial process, which cleans pipes over a period of weeks, did not fit well with the short-term screening used in this study. Although the proprietary product was measured as free chlorine, no inactivation of *Bacillus* spores was observed either in pipe biofilms or as suspended spores.

Decontamination of inorganic compounds was attempted using chlorine, Simple Green, and two chelating agents (sodium citrate and EDTA). Simple Green is a detergent was selected due to its over-the-counter availability and chelating properties. The performance of these agents was marginal, with the highest removals slightly over 50 percent (Tables 14-15). The low level of performance was due, in part, because of the low level of attachment of inorganic contaminants to the pipe surfaces. Most of the compounds attached at levels of 1 mg or less. The sole exception was strontium, which attached to cement-lined ductile iron pipe without a seal-coat and was detected at levels between 14 and 20 mg per pipe segment (Table 14). For these experiments, a 10 percent solution of Simple Green achieved removals between 26 and 56

percent. Because strontium was detected in the cement lining of non-spiked ductile iron pipe segments, it is possible that the performance of the Simple Green cleanser was underestimated due to the release of strontium from the pipe itself. Although the concern for decontaminating inorganic compounds may be low due to their relatively low attachment to the various pipe surfaces (Table 10), radioactive isotopes would be a serious issue because individual atoms would contribute to radioactive decay. This study used non-radioactive salts of cobalt, strontium, cesium, and thallium as surrogates for their radioactive isotopes. The ICPMS detection limit for these compounds ranged between 0.05 and 0.001 mg/L, but concerns over radioactive isotopes may be significant at much lower levels. As indicated in Chapter 3, specialized compounds are used in military applications for decontaminating radioactive surfaces. Additional research should be conducted to evaluate performance goals for decontamination procedures for treatment of radioactive substances.

Initial screening of 11 surfactants, evaluated for color/clarity, foam intensity, and transformation (dissolved status) when introduced into chlordane, resulted in the selection of three surfactants for examination in the decontamination phase. Surfonic N-60 (nonylphenol ethoxylate) is a nonionic surfactant that has an advantage in that it is not affected by water hardness or pH changes, and is considered a medium to low foaming agent but is not very biodegradable (Rosen 1989; Texaco, 2006). Surfonic TDA-6 (ethoxylated tridecyl alcohol) is also a nonionic surfactant used as an agricultural emulsifier. The product has excellent wetting properties, and is somewhat more biodegradable than alkylphenol ethoxylates (Rosen 1989; Texaco, 2006). Empicol LZV, (mon-C12-C18-alkyl sulfate sodium salt) is solid surfactant, widely used in many applications including cosmetics, shampoos and detergents. As an anionic surfactant it has the advantage of being a stable foaming agent, but the disadvantage of being sensitive to minerals, hardness, and pH changes (Rosen 1989; Texaco 2006). In general, all three surfactants were effective for removal of chlordane from cPVC, heavily corroded galvanized iron, and epoxy-coated steel pipes with decontamination rates approaching 90 percent removal (Tables 16-18). These results were very encouraging given that the mass of attached chlordane ranged between 3.1 and 13.5 mg per pipe. Typically, the higher surfactant concentration (5 percent) performed the best. It is not clear what surfactant concentration would be practical in field applications. One requirement of this study was to minimize the carry over of the surfactant into the washed sample so that the detergent would not interfere with the chemical analysis of the contaminants. It was found that washing the pipe with three pipe volumes of water minimized the detergent carry over such that the samples did not foam when shaken. These data suggest that the detergents could be effectively flushed from the distribution system following decontamination of the pipe surfaces.

None of the surfactants tested, however, was particularly effective for removal of p-DCB. The low level of performance could have been due to the relatively poor attachment of the contaminant to the cPVC and the heavily corroded galvanized iron pipe surface. Control (non-decontaminated) values for p-DCB attachment for these surfaces were less than 1 mg (Tables 16-17). However, the mass of p-DCB attached to epoxy-coated steel pipe was more substantial (6.6 mg). Additional research is necessary to examine other surfactants that could be effective for decontamination of p-DCB attached to epoxy coatings. To design specific treatment regimes it is necessary to understand the specific interaction between surface chemistry between a

contaminant and a pipe surface (Rosen 1989). It may be possible to develop general guidelines based on these studies to tailor suites of surfactants that could be used under various conditions.

The use of epoxy-coatings of distribution system pipes in the US is relatively rare (less than 1% of the mains in service)(Kirmeyer et al. 1994), although epoxy coatings are more commonly used in storage tanks and for pipe lining in other countries outside the US. For storage tanks it would be possible to isolate the tank for treatment. In the instance where no chemical decontamination was available, it would be possible to sand-blast the tank surface and install a new coating.

Conclusions

The current study provided an experimental framework by which contaminants, surfaces and decontamination agents could be screened to provide guidance to water utilities for decontamination of water system infrastructure in emergency situations. Chlorine or chlorine dioxide would be capable of inactivating nearly all microbial contaminants providing adequate disinfectant concentrations and contact times could be maintained, and that the pipe material did not exert an unusual disinfectant demand. Most inorganic contaminants did not attach well to the pipe surfaces examined, however the demands for decontamination of radioactive compounds may be much more stringent and protocols for treatment of radioactive isotopes should be further explored. Having access to an inventory of distribution system materials including pipes, valves, gaskets, linings and coatings will be necessary to formulate specific strategies for infrastructure decontamination. Understanding the specific interaction of organic contaminants with pipe surfaces should lead to general guidelines for selection of surfactants. This study identified three surfactants that were effective for chlordane, a contaminant with a high octanol/water partition coefficient. Additional research should be conducted to develop guidelines for other potential contaminants including pesticides and herbicides, petroleum products, and industrial and warfare chemicals.

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